Carbon Nanotubes – Quantum Wires to Artificial Muscles

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Introduction

Carbon nanotube research, whilst still in its infancy, has come a long way since their discovery in 1991 [1]. Considerable characterisation work is still to be done on nanotubes, however this requires a reliable production method of the raw material in sufficient volumes to enable thorough experimental analysis. As part of his fullerene research, fullerenes being of the same carbon family as carbon nanotubes, Wolfgang Kraetschmer made a major breakthrough in synthesis technology in 1990, with the resultant device now bearing his name and being used in nanotechnology research groups around the world.

Fullerene, C_{60}, is a nearly spherical molecule composed of 60 carbon atoms arranged in pentagons and hexagons, like a soccer ball. Fullerenes are molecules with a well determined finite size (0.71nm for C_{60}). Nanotubes, on the other hand, are tiny needle-like structures. A nanotube can be considered as a seamless roll of a graphene sheet, with a typical diameter of 1nm and lengths up to microns. Nanotubes are molecules in their cross section, but along the axis they are infinite solids. Nanotube physics is an interesting superposition of molecular physics in the cross section and solid state physics along the axis.

Synthesis of Carbon Nanotubes

Nanotubes are a special kind of soot particle, found in cases of incomplete combustion of carbon based materials. However, to optimise the amount of nanotubes, and to isolate single walled carbon nanotubes from multi walled carbon nanotubes (concentric layers of tubes stacked inside one another) synthetic synthesis methods have been developed. The most common route utilises the carbon arc method, the same method Kraetschmer used for his mass production of fullerenes. Figure 1 shows a cross sectional view of the Kraetschmer generator.

The essential features are two graphite rods connected to a power supply. The electric arc between the rods evaporates the graphite. The hot carbon plasma collides with cold inert gas and carbon condenses as soot. If the reaction parameters are right (power, gas flow etc) nanotubes are formed, mostly multi-walled nanotubes, however if a catalyst is mixed to the graphite rods the nanotubes are single walled [2]. Common catalysts are particles of iron, nickel, cobalt and alloys thereof [3].

Another effective method for producing nanotubes is by laser ablation of graphite. This involves direct laser evaporation of a composite graphite-transition metal (e.g. Co,

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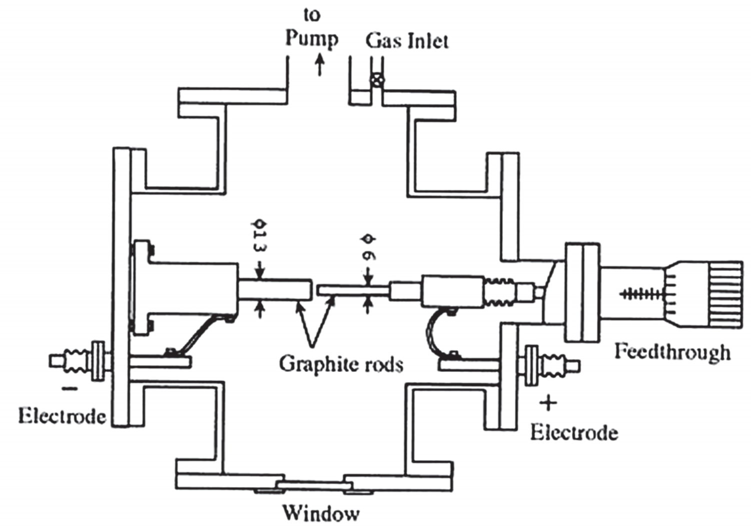

Fig. 1: Schematic of a Kraetschmer generator. (After R. Saito, G. Dresselhaus and M.S. Dresselhaus Physical Properties of Carbon Nanotubes. Imperial College Press, 1998.

Ni) electrode, resulting in a soot, rich in single walled tubes, being deposited in the cooler parts of the reaction chamber [4].

A third process, which has the advantage of being more easily up-scaled to produce large quantities, is thermal decomposition of hydrocarbons, known as chemical vapour deposition (CVD) [5]. CVD can be used to grow bulk phase tubes or templated patterns. Hydrocarbons like acetylene or benzene are heated over catalyst particles and depending on the choice of process parameters, single-walled or multi-walled tubes are obtained or nanofibers and various transition forms between fibers and tubes. A more specific decomposition process, using high pressure and carbon monoxide as the carbon source, results in high purity single walled carbon nanotubes. This method is commonly referred to by the acronym HiPCO (high pressure disproportionation of CO) [6].

Electrical Properties – SWNT and Peapod Transistors

Depending on the helicity of the tube, that is, the angle at which the graphene sheet is rolled, the tube exhibits either metallic or semiconducting behaviour. The electronic character has been the basis for investigating single walled carbon nanotubes as quantum wires for use in electronic device assembly [7]. The underlying physics is that electrical conduction occurs through well separated, discrete electron states that are coherent over the distance between the contacts (typically a few hundred nanometres) and the resistance is independent of the length of the tube (whereas the resistance is proportional to the length for an ordinary wire). There are no losses along the tube (no Joule heating), as the losses occur at the contacts only. This type of charge transport is called ballistic. The conductance along the tube is in integer multiples of the conductance quantum, which is of the order of 100μs. This conductance quantum is a combination of fundamental physical constants, namely the electric charge and Planck's constant: $G_o = e^2/h$. The conductance is therefore a measure for the number of wave guide modes that can propagate in the tube.

Electrical transport of single walled carbon nanotubes has been extensively investigated, revealing fingerprints of ballistic transport, Luttinger-liquid behaviour, and single-electron effects, depending on the details of the experimental set up, and ultimately leading to the application of carbon nanotubes in single molecule field-effect transistors [7]. An extreme case of nanostructuring is illustrated by peapods. 'Peapod' is the term used to describe C_{60} molecules [8] or endohedral metallofullerenes [9] encapsulated within a carbon nanotube. Exciting transport properties are being observed on these new carbon materials.

Experiments carried out at our Institut have indicated that a semiconducting single-walled nanotube doped with Dy@C_{82} indicates temperature dependent electron doping on SWNTs and furthermore, that the semiconducting tube can function as either *p* type or *n* type, depending on the charge transfer [11]. The transistor set up, schematically shown in Figure 2a, enabled observation of a transition from *p* to *n* conduction when the samples were cooled from room temperature to 265K. Upon further cooling to temperatures of about 215K, the samples became metal-like. The conductance spectrum in Figure 2b reveals important information on the doping profile and the quantized charging effect of the peapod transistor system [10].

These results are expected to impact carbon-nanotube based molecular electronics since the metallofullerene functionalisation observed here is more stable than alkaline metal-

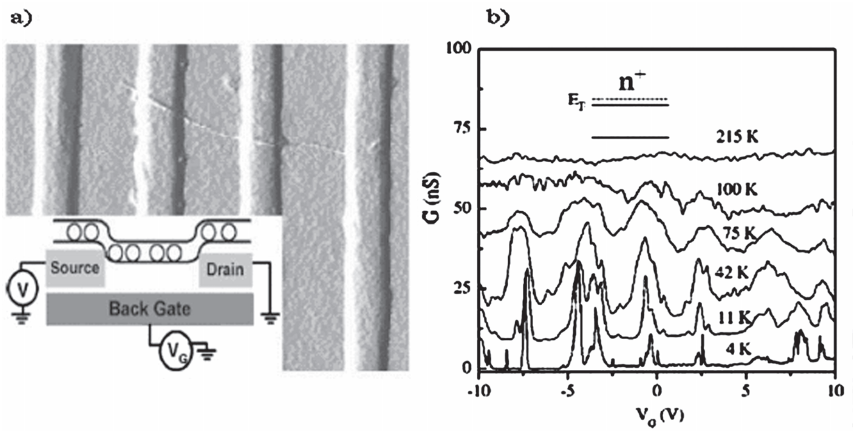

Fig. 2.: a) Atomic force micrograph of individual Dy@C_{82}-SWNT peapod 1.5nm in height. Inset: Schematic of the measurement layout. b) Gate voltage dependence of conductance at a source/drain voltage of 4mV (curves displaced for clarity). Inset: Schematic plot of the band diagram [11].

doped nanotubes [11] in ambient environments. One can envisage an all nanotube transistor, where metallic tubes could function as interconnects [12] and semiconducting tubes can function as field-effect channels [13] or nanorectifiers [14], with the resultant device more of an electromechanical transistor than traditional field-effect transistor.

Mechanical Properties – Nanoactuators from Buckypaper

Carbon nanotubes, with a theoretical Youngs Modulus of 1TPa, are among the strongest solids and can be described as the ultimate carbon fibre. This is important in composite technology, where nanotubes can be incorporated to reinforce the host material. The size and strength of carbon nanotubes also make them ideal candidates for actuating applications, since charge injection into the tube results in the conversion of electrical energy into mechanical energy (actuation) [15]. Actuation has been observed on a macroscale, with entangled single-walled nanotube mats (known as buckypaper) measuring elastic moduli of up to 1.2GPa for an input voltage of only 1-2V [16]. The observed actuation forces are well below those determined for individual nanotube bundles, as the buckypaper is not optimised in terms of nanotube alignment. Nanoscale actuators based on single nanotubes do not have these problems, and research work is underway at our Institut on measuring the actuation of a suspended single-walled tube, both in an electrochemical cell [17] and solid state set-up [18]. The long term goal is to incorporate nanotube actuators in nanodevices, for instance a nanopump as illustrated in Figure 3.

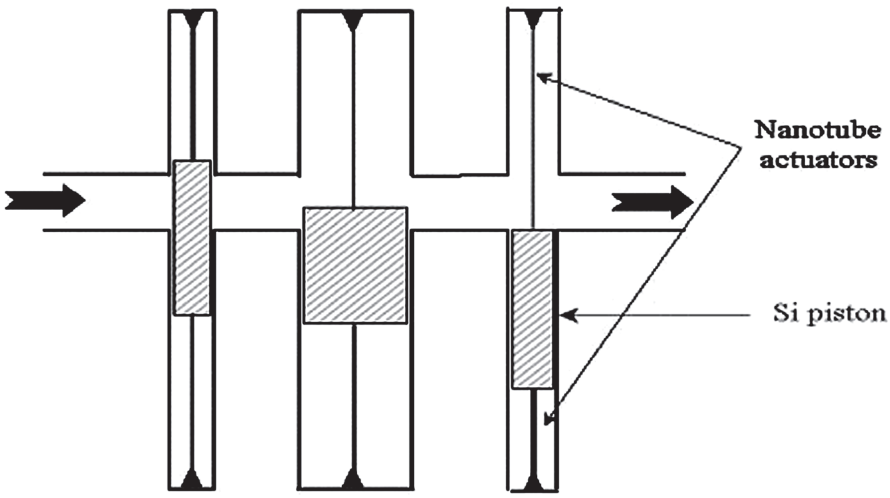

Fig.3: Schematic of a nanoscale pump utilising nanotube actuators as the driving force for the pistons.

However, as described by Baughman [19], for applications outside the nano-realm, individual tubes are too small to exploit their remarkable mechanical properties. A spinning process developed by Vigolo et al produced carbon fibres using unpurified nanotubes (up to 50% impurities), which were measured to have an elastic modulus of up to 15GPa [20]. While this value is substantially lower than the ~600GPa modulus for an individual nanotube [21], it is over an order of magnitude higher than for single-walled buckypaper [22]. It is anticipated that actuation will improve if the spinning process can be modified to generate fibres comprising mostly single-walled carbon nanotubes, and realise applications such as artificial muscles and nanomechanics [23].

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